IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of	Atty. Ref.: **937-1533**
KRAMARZ et al	Conf. No.: **6420**
Serial No. **10/716,920**	Group: **1621**
Filed: **November 19, 2003**	Examiner: **Keys**

For: **PROCESS FOR THE CO-PRODUCTION OF ALCOHOLS**

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December 8, 2008

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

APPLICANTS' BRIEF ON APPEAL

Sir:

This Appeal is from the "final" Official Action dated August 6, 2008, rejecting claims 1, 3-9 and 12-28, all of the claims now pending in this application.[1] As will become evident from the following discussion, the Examiner's rejections are in error and, as such, reversal of the same is solicited.

[1] The claims on appeal, which constitute all presently pending claims in this application, appear in the Section VIII Claims Appendix accompanying this Brief.

I. Real Party In Interest

The real party in interest is the owner of the subject application, namely BASF Corporation.

II. Related Appeals and Interferences

No known appeals and/or interferences related to this application are pending.

III. Status of Claims

A. The following claims are presently pending in this application: Claims 1, 3-9 and 12-28.

B. The following claims are the claims on appeal and have been finally rejected in the Examiner's "final" Official Action of March 6, 2007: Claims 1, 3-9 and 12-28.

C. The following claims have been cancelled during prosecution to date: Claims 2 and 10-11.

D. The following claim(s) have been allowed: None

E. The following claims have been withdrawn: None

F. The following claims have been objected to: None

IV. Status of Amendments

No amendments have been filed subsequent to the “final” Official Action dated August 6, 2008.

V. Summary of Claimed Subject Matter[2]

1. Independent Claim 1

The invention as defined by independent claim 1 is directed to an improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons ([0011]). The improvement comprises enhancing the selectivity of the crossed-aldol condensation reaction through the use of a water-soluble phase-transfer catalyst ([0012]). The phase-transfer catalyst can be removed from the reaction product by water washing ([0013]). The 3-5 carbon aldehyde is propionaldehyde, n-butyraldehyde, isovaleraldehyde, or valeraldehyde ([0011]). The water soluble phase transfer catalyst is a quaternary ammonium or phosphonium salt ([0017]).

2. Independent Claim 3

The subject matter of independent claim 3 is directed to an improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons ([0011]). The improvement comprises enhancing the selectivity of the crossed-aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst ([0017]). The phase-transfer catalyst can be removed from the reaction product by water washing ([0013]). According to the claimed process, 2, 4-diethyloctanol is produced concurrently with 2-ethylhexanol via the hydrogenation of 2-ethyl-2-hexenal and 2,4-diethyl-2-octenal

[2] The numbers in parenthesis refer to the paragraphs of the originally filed specification.

produced from an aldol condensation reaction, which makes use of n-butyraldehyde and 2-ethylhexanal as the reactant aldehydes ([0014], [0016]).

3. Independent Claim 12

The subject matter of independent claim 12 is directed to an improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons ([0011]). The improvement comprises enhancing the selectivity of the crossed-aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst ([0017]). The phase-transfer catalyst can be removed from the reaction product by water washing ([0013]). According to the claimed process, the phase-transfer catalyst is recovered from the water washing by the addition of an alkali metal hydroxide to the water washing to a concentration of 2.5 to 12.5 molar, thereby producing a first phase containing the majority of the phase-transfer catalyst and a second aqueous alkali metal hydroxide phase ([0043]-[0048] and original claim 12).

4. Independent Claim 19

The subject matter of independent claim 19 is directed to an improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons ([0011]). The improvement comprises enhancing the selectivity of the crossed-aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst ([0017]). The phase-transfer catalyst can be removed from the reaction product by water washing

([0013]). According to the claimed process, the aqueous base is the hydroxide form of a quaternary ammonium or phosphonium salt ([0017]).

5. Independent Claim 23

The subject matter of independent claim 23 is directed to an improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons ([0011]). The improvement comprises enhancing the selectivity of the crossed-aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst ([0017]). The phase-transfer catalyst can be removed from the reaction product by water washing ([0013]). According to the claimed process, the crossed-aldol reaction takes place in a three-phase system comprising a first organic aldehyde phase, a second aqueous phase, and a third phase containing the majority of the phase-transfer catalyst ([0017]).

6. Independent Claim 28

The subject matter of independent claim 28 is directed to a process for the preparation of at least one primary alcohol comprising the steps of:

(a) forming an unsaturated aldehyde reaction product by an aqueous base-catalyzed crossed-aldol condensation reaction between a first aldehyde selected from the group consisting of propionaldehyde, n-butyraldehyde, isovaleraldehyde, and valeraldehyde ([0011]) and a second aldehyde containing 6-11 carbons in the presence of a quaternary ammonium or phosphonium salt as a water soluble phase-transfer catalyst (PTC) ([0011}, [0017]), wherein the molar ratio of the PTC to the first aldehyde is between 0.01 to 0.2 ([0021]),

(b) subjecting the unsaturated aldehyde reaction product obtained in step (a) to hydrogenation to form at least one primary alcohol (Figs 2-3, [0015]);

(c) separating the PTC from a process stream containing the same by washing the process stream with water wherein the PTC is recovered (Figs. 2-3, ([0015]); and

(d) recycling the PTC recovered in step (c) in the crossed-aldol condensation reaction of step (a) (Figs. 2-3, ([0015]).

VI. Grounds of Rejection to be Reviewed on Appeal

The following rejection was advanced in the final Official Action dated August 6, 2008:

1. Claims 1, 3-9 and 12-28 stand rejected under 35 USC §103(a) as being unpatentable over Barker et al (USP 4,426,542) in view of Kwok et al (USP 5,801,292), Starks et al (Phase Transfer Catalysts, 1994, pp 482-488), Halpern et al (Spec. Publ.-R. Soc. Chem., 1999, pp 30-39) or Judge et al (UK 1,547,856).

VII. Arguments

1. The Rejection of Claims 1, 3-9 and 12-28 based on the Applied References of Record is in Error.

The Examiner acknowledges that Barker et al "…differ from the instant claims in that Barker et al do not conduct an aldol reaction between mixed aldehydes having different numbers of carbon atoms." (Official action dated January 29, 2008 at page 4, lines 8-9.) To cure this deficiency, the Examiner apparently now turns to Kwok et al. The significant disclosure in Kwok et al appears at column 2, lines 53 through column 3, line 4 and at column 3, line 58 through column 4, lines 11 where it is disclosed that mixed aldehydes may be employed. Specifically, according to Kwok et al, one of the aldehydes may have from two to thirteen carbon atoms (i.e., R_1 in the formula R_1CHO may be an alkyl having one to twelve carbon atoms), while a second aldehyde may have one to thirteen carbon atoms (i.e., R_2 in the formula R_2CHO may be H or an alkyl having one to twelve carbon atoms).

The Examiner then concludes that the mixed aldehydes would "obviously" be employed in the Barker et al process.

The Examiner's analysis is however suggested to be classic hindsight reasoning.[3] Specifically, the Examiner overlooks apparently the admonition in Kwok et al that it is critical that hydrated MgO be employed as the catalyst. ("*A critical aspect of the invention is the use of hydrated MgO for catalyzing or promoting the condensation reaction.*" Col. 4, lines 58-59 of Kwok et al). Thus even though Kwok et al arguably teaches that is it known to have mixed aldehydes in a catalyzed cross-adol reaction, the art is taught that a specific catalyst – namely, hydrated MgO – must critically be

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[3] The Federal Circuit regards hindsight as an insidious and powerful phenomenon and is a tempting, but forbidden zone in the inquiry of addressing the statutory obviousness standard. *See, e.g., Panduit Corp. v. Dennison Mfg. Co.,* 227 USPQ 337 (Fed. Cir. 1985) *and Loctite Corp. v. Ultraseal Ltd.,* 228 USPQ 90, 98 (Fed. Cir. 1985).

employed. Moreover, Kwok et al appear to note that MgO is ***not*** highly water soluble as it may be removed from the reaction zone by centrifuging. (Column 6, lines 38-40.)

Therefore, the ordinarily skilled person having knowledge of Kwok et al would be lead directly away from using mixed aldehydes in a cross-adol reaction in which a water soluble ***quaternary ammonium or phosphonium salt*** was employed as the ***water soluble*** phase transfer catalyst as defined in the applicants' presently pending claims.

The Examiner asserts in response to the argumentation outlined above that:

> "[B]oth Barker et al and Kwok et al aldol condensation reactions, including cross-aldol condensation reactions can be conducted in the presence of other catalyst besides MgO catalysts, for example alkali catalysts are known to catalyze aldol condensation reactions (see column 3, lines 51-53 of Barker et al and column 1, lines 58-65 of Kwok et al)." (August 6, 2008 Official Action at page 4, lines 8-11)

Indeed, the applicants acknowledge that Barker et al disclose at column 3, lines 51-43 that the "...aldol reaction can utilize strongly alkaline catalyst, such as sodium and potassium hydroxide, or sodium and potassium cyanide." And Kwok et al discloses at column 1, lines 58-62 that "...the aldol condensation reaction may be used...for the combination of different aldehydes, producing a so-called "cross aldol", provided that at least one of the aldehydes contains an α hydrogen." However, this disclosure most certainly does ***not*** direct an ordinarily skilled person to a water-soluble PTC generally, or a ***quaternary ammonium or phosphonium salt*** specifically. As noted above, Kwok et al actually instructs the ordinarily skilled person that MgO is ***critically*** required for crossed-aldol reactions.

As such, the disclosure of sodium and potassium hydroxide, or sodium and potassium cyanide cannot be extrapolated erroneously to embrace any and all catalysts – particularly given the unpredictable nature of catalysts generally. In this specific instance, according to the present invention, the use of a water-soluble PTC catalysts, i.e., ***quaternary ammonium or phosphonium salt*** specifically, greater selectivity and yield increases are achieved.

Applicants note that there are at least two major improvements resulting from the use of a water-soluble phase-transfer catalyst as described in paragraph [0008] of the originally filed specification. First, the water soluble PTC improves the solubility of the hydroxide catalyst necessary for the crossed-aldol reaction in the longer chain aldehyde which in turn enhances selectivity for the desired crossed-aldol product versus the self-aldol product produced form the reaction of the shorter chain, more reactive aldehyde. Second, facile recovery of the water soluble PTC is possible from the organic product by aqueous washing.

The secondary references fail to cure the deficiencies of the primary references. In this regard, Judge et al discloses a process using a PTC in a cross-aldol reaction, it is really at this point where the disclosure of Judge et al diverges from the present invention. Specifically, Judge et al fails to teach or suggest using a ***water soluble*** PTC in such a reaction. Applicants note that none of the catalysts listed in the Table at page 4 of Judge et al is considered as "water soluble" by a person skilled in the art.

Starks et al also fail to teach recovering the phase-transfer catalyst from the water washing as correctly stated by the Examiner.

Although Halpern et al mentions the use of Aliquat 100 (tetrabutyl ammonium bromide) and Aliquat 175 (methyl tributyl ammonium chloride) -- which are water soluble PTC catalysts – Halpern et al do not teach any specific use of these catalyst in a

crossed-aldol reaction as claimed in the present application. As noted previously, a significant improvement of the present invention is enhancing the selectivity of the crossed-aldol condensation reaction through the use of a water-soluble phase transfer catalyst.

Thus, while water-soluble phase transfer catalysts may be known per se (e.g., as evidenced by Halpern et al) there is no motivation for a person skilled in the art to combine the use of such water soluble PTC catalyst in an aldol reaction as described by Judge. One skilled in this art simply would not disregard the teaching of Judge et al with respect to use of the non- or only minimally water soluble PTC of Judge et al and substitute therefore a fully water soluble PTC as taught by the present applicants.

Moreover, applicant notes that comparison of the data in the Examples of the subject application (which employs a water soluble PTC in accordance with the present invention) with Comparative Example 2 (which employs a PTC catalyst in accordance with Judge et al, namely tricaprylmethylammonium chloride). As demonstrated by such data, two yield improvements are evident – namely the total conversion of n-butyraldehyde to both 2,4-diethyl-2-octenal and 2-ethyl-2-hexenal and a higher yield increase (i.e., greater selectivity) for 2,4,-diethyl-2-octenal.

The yield increases demonstrated by the data in the Examples of the subject application are significant in commercial terms. Specifically, on a commercial scale, the US production capacity of 2-ethylhexanol is approximately 400,000 metric tons per year, with some individual production plants having production capacities of 130,000 metric tons per year. Since 2,4-diethyloctanol is a higher molecular weight alcohol than 2-ethylhexanol and the plasticizer formed from 2,4-diethyloctanol (i.e., bis(2,4-diethyloctyl)phthalate) has superior plasticizer properties as compared to the plasticizer formed from the lower molecular weight 2-ethylhexanol (i.e., di(2-ethylhexyl)phthalate), the former higher molecular weight alcohol commands significant price premium. Thus,

on a large scale production plants, even seemingly modest yield improvements result in significant realized income for the producer. Thus, the yield improvements noted in the Examples of the subject application using a water soluble PTC in accordance with the present invention in comparison to Comparative Example 2 using the catalyst in accordance with Judge et al is quite significant technically.

2. Conclusion.

In view of the above, applicants submit that the present invention is patentably *un*obvious over the applied references of record. For the reason advanced, the Examiner's rejection of the pending claims herein under 35 USC §103(a) is in error and must be reversed. Such favorable action is solicited.

Respectfully submitted,

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VIII. CLAIMS APPENDIX

1. (previously presented) An improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons, the improvement comprising enhancing the selectivity of the crossed-aldol condensation reaction through the use of a water-soluble phase-transfer catalyst, and removing the phase-transfer catalyst from the reaction product by water washing, wherein the 3-5 carbon aldehyde is propionaldehyde, n-butyraldehyde, isovaleraldehyde, or valeraldehyde, and wherein the water soluble phase transfer catalyst is a quaternary ammonium or phosphonium salt.

2. (canceled)

3. (previously presented) An improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons, the improvement comprising enhancing the selectivity of the crossed-aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst, and removing the phase-transfer catalyst from the reaction product by water washing, wherein 2, 4-diethyloctanol is produced concurrently with 2-ethylhexanol via the hydrogenation of 2-ethyl-2-hexenal and 2,4-diethyl-2-octenal produced from an aldol condensation reaction, which makes use of n-butyraldehyde and 2-ethylhexanal as the reactant aldehydes.

4. (original) The process of claim 3, wherein the molar ratio of 2-ethylhexanal to n-butyraldehyde fed to the crossed-aldol condensation reaction is about 1 to about 5.

5. (original) The process of claim 3, wherein the molar ratio of 2-ethylhexanal to n-butyraldehyde fed to the crossed-aldol condensation reaction is about 1 to about 10.

6. (previously presented) The process of claim 3, wherein said 2-ethylhexanal is produced by partial hydrogenation of 2-ethyl-2-hexenal using a Group VIII metal catalyst, said 2-ethyl-2-hexenal being produced by an aldol condensation reaction of n-butyraldehyde.

7. (previously presented) The process of claim 3, wherein a portion of the unreacted 2-ethylhexanal and 2-ethyl-2-hexenal are recovered from the crossed-aldol condensation reaction product in preference to hydrogenation to 2-ethylhexanol.

8. (original) The process of claim 7, wherein 2-ethylhexanal is produced by the Group VIII metal catalyzed partial hydrogenation of said recovered 2-ethyl-2-hexenal.

9. (original) The process of claim 8, wherein the Group VIII metal is palladium.

10. (cancelled)

11. (cancelled)

12. (previously presented) An improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons, the improvement comprising enhancing the selectivity of the crossed-

aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst, and removing the phase-transfer catalyst from the reaction product by water washing, wherein the phase-transfer catalyst is recovered from the water washing by the addition of an alkali metal hydroxide to the water washing to a concentration of 2.5 to 12.5 molar, thereby producing a first phase containing the majority of the phase-transfer catalyst and a second aqueous alkali metal hydroxide phase.

13. (original) The process of claim 12, wherein the alkali metal hydroxide is sodium hydroxide.

14. (previously presented) The process of claim 12, wherein the phase-transfer catalyst has a cationic portion which is methyltributylammonium, tetrabutylammonium, benzyltriethylammonium, ethyltributylammonium, tetraethylammonium, tetrahexylammonium, tetrapropylammonium, or tetrabutylphosphonium.

15. (previously presented) The process of claim 12, wherein the phase-transfer catalyst has an anionic portion which is chloride, bromide, iodide, bisulfate, sulfate, or hydroxide.

16. (original) The process of claim 1, wherein the aqueous base is an alkali metal hydroxide.

17. (original) The process of claim 16, wherein the alkali metal hydroxide is sodium hydroxide or potassium hydroxide.

18. (previously presented) The process of claim 17 wherein the aqueous base comprises a 10-50 weight percent solution of sodium hydroxide.

19. (previously presented) An improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons, the improvement comprising enhancing the selectivity of the crossed-aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst, and removing the phase-transfer catalyst from the reaction product by water washing, wherein the aqueous base is the hydroxide form of a quaternary ammonium or phosphonium salt.

20. (original) The process of claim 1, wherein the aldol reaction is performed at a temperature from about 30 to 100^{0}C.

21. (original) The process of claim 1, wherein the aldol reaction is performed at a temperature from about 30 to about 120^{0}C.

22. (original) The process of claim 1, wherein the crossed-aldol reaction takes place in a two-phase system comprising a first organic aldehyde phase and a second aqueous phase, and the phase-transfer catalyst is primarily in the organic aldehyde phase.

23. (previously presented) An improved process for the preparation of at least one primary alcohol by the hydrogenation of an unsaturated aldehyde reaction product produced by an aqueous base-catalyzed crossed-aldol reaction between a first aldehyde containing 3-5 carbons and a second aldehyde containing 6-11 carbons, the improvement comprising enhancing the selectivity of the crossed-aldol condensation reaction through the use of a quaternary ammonium or phosphonium salt as a water-soluble phase-transfer catalyst, and removing the phase-transfer catalyst from the reaction product by water washing, wherein the

crossed-aldol reaction takes place in a three-phase system comprising a first organic aldehyde phase, a second aqueous phase, and a third phase containing the majority of the phase-transfer catalyst.

24. (previously presented) The process of claim 1, wherein the molar ratio of the phase-transfer catalyst to the first aldehyde is about 0.01 to about 1.

25. (original) The process of claim 1, wherein the molar ratio of aqueous base to the first aldehyde is about 0.1 to about 2.

26. (original) The process of claim 1, wherein the aldol reaction is performed in a continuous or batch reactor.

27. (original) The process of claim 1, wherein the unsaturated aldehyde reaction product is hydrogenated in the gas and/or liquid phase in a single or multistage process.

28. (previously presented) A process for the preparation of at least one primary alcohol comprising the steps of:
    (a) forming an unsaturated aldehyde reaction product by an aqueous base-catalyzed crossed-aldol condensation reaction between a first aldehyde selected from the group consisting of propionaldehyde, n-butyraldehyde, isovaleraldehyde, and valeraldehyde and a second aldehyde containing 6-11 carbons in the presence of a quaternary ammonium or phosphonium salt as a water soluble phase-transfer catalyst (PTC), wherein the molar ratio of the PTC to the first aldehyde is between 0.01 to 0.2,
    (b) subjecting the unsaturated aldehyde reaction product obtained in step (a) to hydrogenation to form at least one primary alcohol;

(c) separating the PTC from a process stream containing the same by washing the process stream with water wherein the PTC is recovered; and

(d) recycling the PTC recovered in step (c) in the crossed-aldol condensation reaction of step (a).

IX. EVIDENCE APPENDIX

[NONE]

X. RELATED PROCEEDINGS APPENDIX

[NONE]